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Receptor Models Relating Ambient Suspended Particulate Matter to Sources

Environmental Research and Technology, Inc.
Concord, MA

Prepared for

Industrial Environmental Research Lab.
Research Triangle Park, NC

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RECEPTOR MODELS RELATING AMBIENT SUSPENDED PARTICULATE MATTER TO SOURCES

Prepared for

Office of Air Quality Planning and Standards

Prepared by

Industrial Environmental Research
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Research Triangle Park NC 27711

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16. ABSTRACT The report describes the use of receptor models to determine the source contributions to ambient particulate matter loadings at sampling sites, based on common properties between sources and receptors. (This is in contrast to using source models which start with emission rates and meteorological measurements to predict ambient concentrations.) Three generic receptor models have been identified: chemical mass balance, multivariate, and microscopial identification. Each has certain requirements for input data to provide a specific output. An approach that combined receptor and source models, source/receptor model hybridization, has also been proposed, but it needs further study. The input to receptor models is obtained from ambient sampling, source sampling, and sample analysis. The design of the experiment is important in obtaining the most information for the least cost. Sampling schedule, sample duration, and particle sizing are part of the ambient sampling design. Analysis for elements, ions, carbon, and organic and inorganic compounds is included in the sample analysis design. Which sources to sample and how to sample them are part of the source sampling design.

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RECEPTOR MODELS RELATING AMBIENT SUSPENDED PARTICULATE MATTER TO SOURCES

Prepared by

John G. Watson
Environmental Research and Technology, Inc.
696 Virginia Road
Concord, MA 01742

EPA Project Officer

John O. Milliken

Contract No. 68-02-2542, Task No. 8

Prepared for

U.S. Environmental Protection Agency
Office of Research and Development
Washington, D.C. 20460

Prepared by

John G. Watson

with assistance from

William Baasel
John Bachman
Neil Berg
Glenn Cass
John Cooper
John Core
Russell Crutcher
Lloyd Currie
Joan Daisey
Stuart Dattner
Briant Davis
Ronald Draftz
Alan Dunker
Thomas Dzubay
Robert Eldred
Edward Fasiska
Sheldon Friedlander
Donald Gatz
Glen Gordon
Bruce Harris
Ronald Henry
George Hidy
Philip Hopke
Reginald Jordan
Ted Kneip
Nick Kolak
Ross Leadbetter
Richard Lee
Charles Lewis
Carol Lyons
John Milliken
Jarvis Moyers
John Overton
Thompson Pace
Skip Palenik
Terry Peterson
William Pierson
Kenneth Rahn
Philip Russell
John Spengler
Robert K. Stevens
Warren White
Jack Winchester
John Woodward
John Yocom

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This report is the product of many minds, as evidenced by the authorship on the title page.

It results from a two and one half day meeting in which these researchers and users of receptor models came together in formal and informal sessions at the Quail's Roost Conference Center of the University of North Carolina located in Rougemont, North Carolina. The intimate, tranquil setting and the southern hospitality of the Quail's Roost was a major ingredient in the success of the meeting, and the staff, under the leadership of Mr. Charlie Philips, merits special appreciation.

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The final acknowledgement goes to those pioneers of receptor modeling who did not participate in the Quail Roost meeting but upon whose efforts the science is being developed. As much as possible, relevant references to their work have been included in the bibliography.

ABSTRACT

Receptor Models are used to determine the source contributions to ambient particulate matter loadings at a sampling site based on common properties between source and receptor. This is in contrast to a source model which starts with emission rates and meteorological measurements to predict an ambient concentration.

Three generic types of receptor model have been identified, chemical mass balance, multivariate, and microscopical identification. Each one has certain requirements for input data to provide a specified output. An approach which combines receptor and source models, source/receptor model hybridization, has also been proposed, but it needs further study.

The input to receptor models is obtained from ambient sampling, source sampling, and sample analysis. The design of the experiment is important to obtain the most information for least cost. Sampling schedule, sample duration and particle sizing are part of the ambient sampling design. Analysis for elements, ions, carbon, organic and inorganic compounds are included in the sample analysis design. Which sources to sample and how to sample them are part of the source sampling design.

In order for receptor modeling to become a useful tool, it must be developed in five major areas:

- General Theory - The generic types of receptor model are related to each other, but that relationship has not been generally established. A general theory of receptor models, including the input data uncertainties, needs to be constructed.
- Validation - Simulated data sets created from known source contributors and perturbed by random error need to be presented to models, and their source contribution predictions should be compared to the known contributions. Several models need to be applied to the same data set and their results compared.
- Standardization - Validated models need to be placed in standard form for easy implementation and use.

- Measurement Methods - Ambient and source sampling and analysis methods need to be developed, tested and standardized.

- Documentation and Education - Users need to be informed of the powers and limitations of these models and instructed in their use.

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CHAPTER 1: INTRODUCTION

There is a need today to quantify the effects of aerosol sources on ambient particulate matter loadings. Over one hundred air quality maintenance areas in the United States are not meeting primary and secondary Total Suspended Particulate (TSP) Matter Standards. These areas must submit state implementation plans showing how they will come into attainment within a specified time period. This involves the identification of major causes of 24 hour and/or annual standard violations and the application of a control strategy to eliminate those causes. The source identification step rarely receives as much attention as the control of "assumed" sources. The result of this imbalance can result in expensive controls with little or no effect on ambient air quality. Industry is reluctant to make expenditures when it sees no hard evidence that it is a major contributor to violations. Airshed residents balk at higher taxes and burning and driving restrictions in the absence of a clear relationship between those activities and air quality.

As the condition of the environment improves, people become concerned about certain amenities in addition to health effects. Industrial sources are often charged as the cause of soiling, deposition or visibility degradation in a community or area. The verification of these charges is often left to emotional pleas rather than to hard technical evidence. When an industry admits culpability, it still needs to determine which part of its process is the major contributor.

Standards for suspended particulate matter must be reevaluated every ten years. At the present time the Environmental Protection Agency is proposing a size-selective aerosol standard (Miller, et al, 1979). The economic and monitoring impacts of such a standard cannot be assessed without determining relative source contributions to the size-range under consideration.

Identifying the major sources of ambient particulate matter loadings was a fairly simple process when values exceeded 500 $\mu g/m^3$ and stack emissions were plainly visible. Control of these emitters was forthcoming and effective. At levels of 150 to 200 $\mu g/m^3$, the use of annual emission inventories focused further regulatory efforts on major sources which have resulted in more successful reductions.

Presently, at levels around 75-100 $\mu g/m^3$, the uncertainties involved in these assessments of source contributions are greater than the contributions themselves.

Source-oriented atmospheric dispersion modeling has been the major tool used in attributing ambient concentrations to source emissions. With extensively researched emission inventories and adequate meteorological information, these models have done a credible job of predicting non-reacting gaseous pollutant concentrations at receptors due to specific sources. Many times, however, predicted concentrations do not compare well with ambient measurements (e.g., Guldberg and Kern, 1978).

These source models have not proved themselves adequate for suspended particulate matter for several reasons. First, particulate matter emissions are widely dispersed and hard to quantify. Whereas most gaseous emissions can be confined to specific points (even auto tailpipes, though numbering in the millions, are identifiable and confined to roadways and parking lots), particulate matter is produced from point and nonpoint sources; every square inch of surface crossed by wind is a potential emitter. Emission factors for area sources are hard to estimate and display wide variability.

Second, the transport of aerosol is highly dependent on particle size, with larger particles settling out closer to the emission point than smaller ones or impacting out on surfaces such as trees or buildings. The particle size distributions of many aerosol sources are inadequately characterized or highly variable. Even if the distributions were available, few source models have developed mechanisms for aerosol removal. There are source models in which total reflection at ground level occurs!

Finally, the complexities of aerosol chemistry, -- the condensation and adsorption of organic vapors -- the conversion of SO_2 to sulfate species -- is not a part of the standard source models. Though these models may potentially provide useful insights, their results must not be relied upon as the definitive statement.

With the development of inexpensive and rapid chemical analysis techniques for dividing ambient and source particulate matter into its components has come another approach, the receptor model.

While the source-oriented model begins with measurements at the source (i.e., emission rates for the period under study), and estimates ambient concentrations, the receptor-oriented model begins with the actual ambient measurements and estimates the source contributions to them. Figure 1 illustrates the difference between source and receptor models.

The receptor model relies on properties of the aerosol which are common to source and receptor and that are unique to specific source types. These properties are composition, size and variability.

The composition of material from one source type is different from that of another. Auto exhaust particles contain significant amounts of lead and bromine not found in aerosol from other sources. The shapes, birefringences and x-ray diffraction patterns of certain minerals can identify which aggregate storage pile they come from if the piles contain different minerals.

The particle size distribution is dictated by the processes which generate the aerosol, the most useful size distinction being between particles greater than and less than 2 μm. Willeke and Whitby (1975), after hundreds of particle size measurements in urban situations, have found that the mass of most aerosols appears to be concentrated into two size ranges, commonly termed the coarse (particles >2 μm) and fine (particles <2 μm) particle modes. Coarse particles are primarily the result of grinding and errosive operations, the breaking up of larger material into smaller pieces. Geological material tends to dominate this mode. Fine particles are formed by the condensation of gases to particles, chemical reactions between gases yielding particles, and the coagulation of still smaller

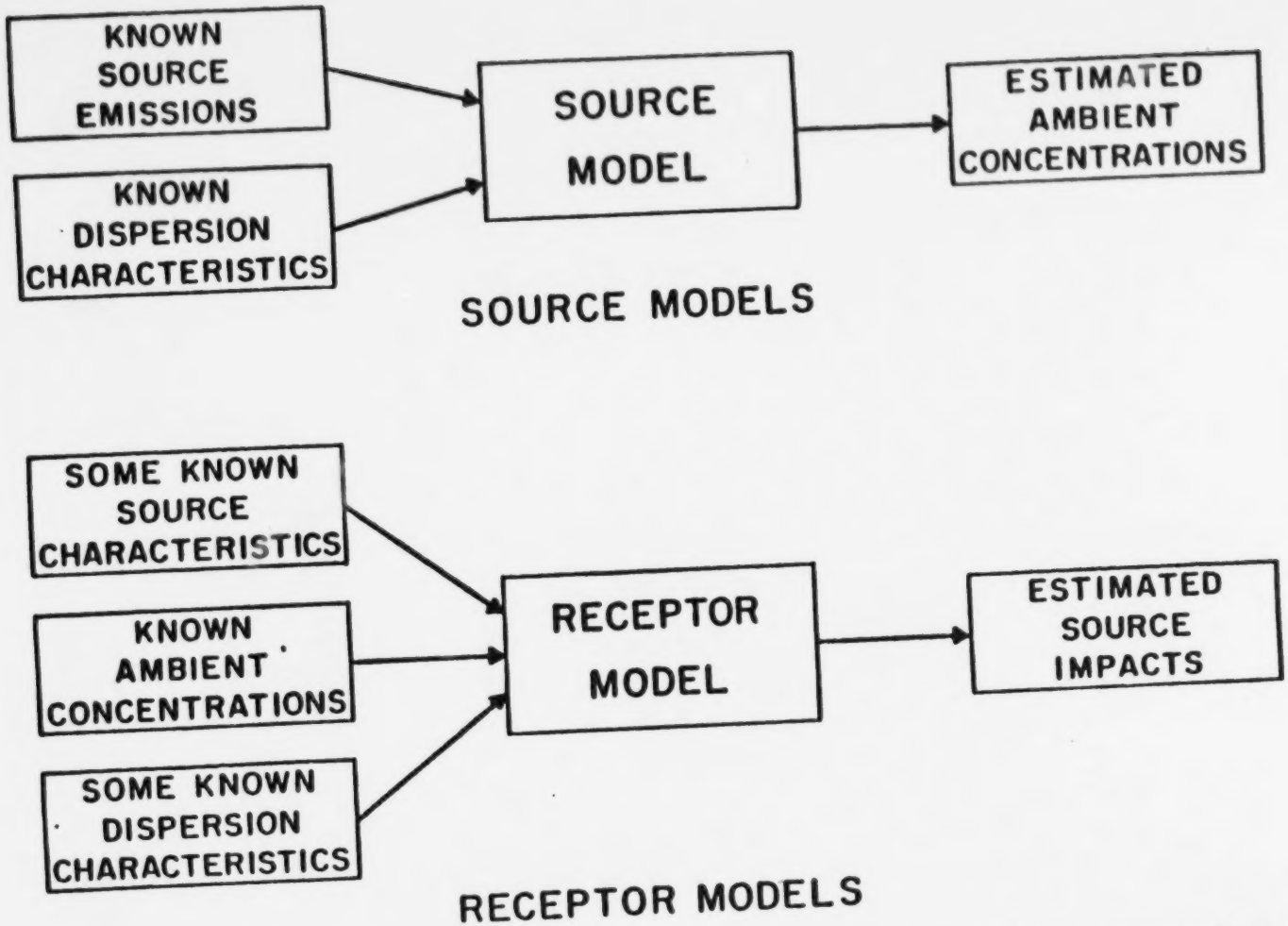

Figure 1. The source model uses source emissions as inputs, calculates ambient concentrations. The receptor model uses ambient concentrations as inputs, calculates source contributions. (From Watson, 1979)

particles. A dichotomous mass sample, the measurement of the fraction of the total mass contributed by the coarse and fine modes, is a good indicator (Dzubay and Stevens, 1975) of the relative contributions from these two broad source categories. It appears (Heisler et al., 1973) that finer gradations of particle size at source and receptor could make these classifications more specific.

The variation of particulate matter properties, both temporally and spatially, corresponds to variations of source emissions and to the variability of meteorological transport between source and receptor. Increases in ambient lead and bromine concentrations during rush hour could correspond to a known increase in auto exhaust during those time periods. Rheingrover and Gordon (1980) show spatial variability of aerosol component concentrations varying precisely with wind direction, implicating upwind sources known to emit those components. When the effects of changing meteorology are taken into account, sources with diurnal, weekly, and seasonal emission variations will engender similar variations of the components they contribute to a receptor. Source/receptor distance and wind related transport cause all source contributions to vary with receptor location.

The examination of these properties to identify source contributions is not new. Throgmorton and Axetell (1978) have done a credible job of tabulating and generalizing applications of receptor models in the past and Cooper and Watson (1980) and Gordon (1980) offer excellent reviews of current work. Receptor modeling is an active area of research which is on the verge of becoming a useful tool in the ambient monitoring and regulatory process. However, it needs a unified focus to direct its development.

In an attempt to provide this focus, forty-seven active receptor model users from government, university, consulting and industry met for 2 1/2 days in February 1980. They addressed the models and the information required to use them in six separate task forces:

- Chemical Element Balance Receptor Models
- Multivariate Receptor Models

- Microscopic Identification Receptor Models
- Field Study Design and Data Management
- Source Characterization
- Analytical Methods

The objectives of these inter-related task forces were to:

- define the state-of-the-art via summaries of past and present research,
- outline future research required to make the models useful tools for the scientific community, regulators, and planners,
- suggest vehicles for carrying out that research within the context of ongoing projects.

This report presents the results of this meeting with sufficient background material and references on receptor models to make the recommendations meaningful to uninitiated but potential receptor model users.

This introduction presents the general concept of a receptor model, the input it requires, and the output it produces. Chapter Two will describe chemical mass balance, multivariate, microscopic and hybrid model approaches, the specific data they require, the recognized state-of-the-art, and research directions for each model.

Having specified the data required in Chapter Two, Chapter Three will summarize the state-of-the-art of the methods of obtaining those data in terms of field study design, source characterization, and analytical methods. Areas requiring exploration and refinement will be stated.

Chapter Four makes specific recommendations for meeting the needs proposed in Chapters Two and Three.

The extensive bibliography following Chapter Four, and the liberal reference to it thoughout the text, is intended to provide direction to present and future researchers and users of receptor modeling methods.

CHAPTER 2: THE RECEPTOR MODELS

Receptor models presently in use can be classified into one of four categories: chemical mass balance, multivariate, microscopic, and source/receptor hybrids. Each classification will be treated individually, though it will become apparent that they are closely related.

The starting point for the receptor model is the source model. Though the source model may not deliver accurate results under many conditions, its limitations are primarily due to its inability to include every environmentally relevant variable and inadequate measurements for the variables it does include. The general mathematical formulations, however, are representative of the way in which particulate matter travels from source to receptor.

In general, the source model states that the contribution of source j to a receptor, S_j, is the product of an emissions rate, E_j, and a dispersion factor, D_j,

$$S_j = D_j E_j \tag{1}$$

Various forms for D_j have been proposed (Pasquill, 1974, Benarie, 1976, Seinfeld, 1975), some including provisions for chemical reactions, removal, and specialized topography. None are completely adequate to describe the complicated, random nature of particulate matter travel in the atmosphere. Similarly, E_j is difficult to quantify on an absolute basis for many sources. The advantage of the receptor model is that exact knowledge of D_j and E_j is unnecessary.

If a number of sources, p, exists and there is no interaction between their aerosols to cause mass removal, the total aerosol mass measured at the receptor, C, will be a linear sum of the contributions of the individual sources.

$$C = \sum_{j=1}^{p} S_j \tag{2}$$

Similarly, the mass concentration of aerosol property i, C_i, will be

$$C_i = \sum_{j=1}^{p} a_{ij} S_j \qquad (3)$$

where a_{ij} is the mass fraction of source contribution j possessing property i at the receptor.

2.1 The Chemical Mass Balance Receptor Model

Equation 3 looks similar to Friedlander's (1973) chemical element balance which has been used in Pasadena (Miller et al. 1972, Hammerle and Pierson, 1975), Chicago (Gatz, 1975), Fresno, Pomona, San Jose, Riverside (Gartrell and Friedlander 1975), New York (Kneip et al 1972, Daisey et al., 1979), Portland (Henry, 1977, Watson, 1979), Washington, D.C. (Kowalczyk et al. 1978), Denver (Heisler et al. 1980, Courtney et al., 1980), Miami (Hardy, 1979), Smokey Mountains (Stevens et al. 1980), and St. Louis (Dzubay et al 1980) to assess source contributions. If the p sources are considered "source types", that is, grouping sources with similar properties together, the set of equations like Equation 3 for all of the elemental concentrations is the chemical element balance.

A better name for this model might be chemical mass balance. Historically, elemental measurements were most readily available. Recent analytical developments allow chemical compounds and ions to be among the properties considered.

If one measures n chemical properties of both source and receptor, n equations of the form of Equation 3 exist. If the number of source types contributing those properties is less than or equal to the number of equations, i.e., $p \leq n$, then the source contributions, the S_j, can be calculated.

Four methods of performing this calculation have been proposed, the tracer property, linear programming, ordinary linear least squares fitting, and effective variance least squares fitting.

The tracer element method is the simplest. It assumes that each aerosol source type possesses a unique property which is common to no other source type, Equation 3 then reduces to

$$S_j = \frac{C_{t_j}}{a_{t_j j}} \qquad (4)$$

for each source j with its tracer t_j. It works well when the tracers meet the following requirements:

1. $a_{t_j j}$ perceived at the receptor is well known and invariant.
2. C_{t_j} can be measured accurately and precisely in the ambient sample.
3. The concentration of property t_j at the receptor comes only from source type j.

These conditions cannot be completely met in practice, and by limiting the model to only one tracer property per source type, valuable information contained in the other aerosol properties is being discarded. Thus, solutions to the set of equations like Equation 3 have been developed to make use of the additional information provided by more than one unique chemical property of a source type, and even that of properties which are not so unique.

The linear programming and weighted least squares solutions deal with the case of a number of constraints, n, greater than the number of unknowns, p.

Henry (1977) has applied a linear programming algorithm (Hadley, 1962) which maximizes the sum of the source contributions subject to the following constraints:

$$0 \leq S_j \leq C \tag{5}$$

$$C_i + 3\sigma_{C_i} \leq \sum_{j=1}^{p} a_{ij} S_j \tag{6}$$

$$0 \leq \sum_{j=1}^{p} S_j + 3\sigma_C \tag{7}$$

where σ_{C_i} is the uncertainty in the measurement C_i.

This solution has not developed further.

In the ordinary weighted least squares method, the most probable values of S_j when $n > p$ are achieved by minimizing the weighted sum of squares of the difference between the measured values of C_i and those calculated from Equation 3 weighted by σ_{C_i}, the analytical uncertainty of the C_i measurement.

$$\chi^2 = \sum_{i=1}^{n} \frac{\left(C_i - \sum_{j=1}^{p} a_{ij} S_j \right)^2}{\sigma_{C_i}^2} \tag{8}$$

This solution provides the added benefit of being able to propagate the measured uncertainty of the C_i through the calculations to come up with a confidence interval around the calculated S_j.

The ordinary weighted least squares solution is incomplete, however. The ambient aerosol chemical properties, the C_i, are not the only observables on which measurements are made. The chemical properties of the source aerosol, the a_{ij}, are also measured observables, but the errors associated with those measurements are not included in the ordinary weighted least squares fit.

Dunker (1979) and Watson (1979) have applied the treatment of Britt and Luecke (1973) to the solution of a set of Equations 3. The maximum likelihood solution minimizes the function

$$\chi^2 = \sum_{i=1}^{n} \frac{\left(C_i - \sum_{j=1}^{p} a_{ij} S_j \right)^2}{\sigma_{C_i}^2 + \sum_{j=1}^{p} \sigma_{a_{ij}}^2 S_j^2} \qquad (9)$$

where $\sigma_{a_{ij}}$ is the uncertainty associated with the a_{ij} measurement.

This solution provides two benefits. First, it propagates a confidence interval around the calculated S_j which reflects the cumulative uncertainty of the input observables. The more precise the measurements of the ambient source property concentrations are, the better the estimate of the source contributions will be. The second benefit provided by this "effective variance" weighting is to give those chemical properties with larger uncertainties, or chemical properties which are not as unique to a source type, less weight in the fitting procedure than those properties having more precise measurements or a truly unique source character.

In a series of simulation studies on artifically generated, randomly perturbed data sets, Watson (1979) shows that these benefits are indeed realized in practice.

Each least squares solution exhibits instabilities when source compositions are similar, though not exact. Very large or negative source contributions result. Other fitting methods such as constrained least squares (Twomey, 1977) and non-negative least squares (Leggett, 1977) need to be explored with the addition of the uncertainties in the source compositions.

The input data required for the chemical mass balance consist of the following:

1. The concentrations of the chemical components which comprise the total mass of the sample, the C_i, and their precisions, the σ_{C_i}.

2. The number, p, and identification of source types contributing to the receptor samples.

3. The source type compositions, the a_{ij}, and their variabilities $\sigma_{a_{ij}}$.

Obtaining this input data is not a trivial matter. The greatest progress in recent years has been made in obtaining the data in requirement 1, the ambient chemical component concentrations. Chapter Three will present the most widely used sampling and analytical tools.

Meeting the second data requirement is more difficult. In fact, one of the objectives of the receptor model must be to identify as well as to quantify source contributions. The maximum number of source types affecting a sampler in a given airshed will surely correspond to the ones located within that airshed. Some, such as geological material, automobile exhaust, oil and wood combustion are located throughout the airshed and might be included first. Gaps between the calculated and measured aerosol property concentrations suggest other sources, which are included in a new calculation, yielding new gaps, suggesting new sources, etc. Watson (1979) tried this in Portland. Although the results are reasonable, they are also subjective. A method of identifying the sources in the mass balance needs to be devised.

A variation of the chemical mass balance which has been used to identify additional source types is the enrichment factor defined as

$$E_i = \frac{C_i / C_t}{a_{ib} / a_{tb}} \tag{10}$$

where the b subscript refers to a dominant source type contribution, usually crustal material, and the t subscript identifies a tracer element, such as Al or Fe, unique to that source type. Substituting Equations (3) and (4) into Equation 10 yields

$$E = 1 + \frac{\sum_{j \neq b}^{p} a_{ij} S_j}{a_{ib} S_b} \tag{11}$$

If no other sources of chemical component i exist, $E_i = 1$. E_i greater than one implies sources of component i other than source b. For example, King et al. (1976) noticed a high Sb enrichment at one site in Cleveland, Ohio. Further investigation revealed the existence of a nearby chemical plant producing Sb compounds.

Ambient aerosol property measurements are quite sophisticated; the state of source measurements is atrocious. Certain aerosol property measurements have been made on emissions from geological material (Rahn, 1976), auto exhaust (Pierson and Brachaczek, 1974), marine background (Hidy et al. 1974), coal combustion (Gladney et al. 1976), municipal incinerators (Greenberg, 1978), iron and steel blast furnaces (Jacko, 1977), tire dust (Pierson and Brachaczek, 1974), copper smelters (Zoller et al. 1978), vegetative burning (Core and Terraglio, 1978), kraft recovery boilers (Watson, 1979) and cement plants. The references cited are representative rather than exhaustive. These tests seldom provide a full accounting of aerosol properties, have not taken samples similar to the ambient samples, report no confidence intervals on the measurements, and do not fully describe the operating parameters of the source at the time of the test.

Even if a methodology existed for exactly quantifying the source type compositions, it is unlikely that all individual emitters would have the same composition or even that the composition would remain the same over the period of time during which ambient samples are taken.

The source contributions of aerosol formed from gaseous emissions, such as sulfate, nitrate and certain organic species, cannot be quantified by chemical mass balance methods. Watson (1979) proposes a unique source type which will put an upper limit on the contributions of secondary aerosol sources, but it cannot attribute those contributions to specific emitters.

Even given these limitations, users of the chemical mass balance feel that the method has been developed to the point where it can frequently be used as a basis for state implementation plans for the control of particulate matter pollution. It is conceptually simple, based on real data, and given the proper input data, it is able to

quantitatively apportion contributions of sources to ambient air quality. The difference between the mass concentration at the receptor and the sum of the source contributions offers a consistency check; if the sum is much greater than the total mass, one or more of the source contributions have been over-estimated; if the sum is much less than the mass, sources have been left out or some of those included do not belong. This same check can be applied to chemical components not included in the fitting procedure.

Major source contributions are quantified with greater precision than minor contributions, but this may not be a limiting feature for many purposes. Watson (1979) observed that the industrial source contributions to the Portland airshed were subject to large uncertainties; if their exact determination was required, the results would have been inadequate. However, for the control-strategy-planning purposes of that study, they were sufficient for concluding that the maximum possible industrial contributions were not causing standards violations.

The future development of the chemical mass balance receptor model must include:

- More chemical components measured in different size ranges at both source and receptor.
- Study of other mathematical methods of solving the chemical mass balance equations.
- Validated and documented computer routines for calculations and error estimates.
- Extension of the chemical mass balance to an "aerosol properties balance" to apportion other aerosol indices such as light extinction.

2.2 Multivariate Models

The chemical mass balance uses the aerosol property of chemical composition. If source and ambient samples are taken in more than one size range, the size property can be used to separate the contributions

of one source from another. The mass balance presently has no way to incorporate the variability of ambient concentrations and source emissions. The multivariate methods can do this.

Linear regression (Kleinman, 1977, 1980, Hammerle and Pierson, 1975, Neustadter et al., 1976), correlation (Moyers et al., 1977, Cahill et al., 1977) and factor analysis (Hopke et al., 1976, Henry, 1977) are the forms these models take.

While the chemical mass balance receptor model was easily derivable from the source model and the elements of its solution system were fairly easy to present, this is not the case for multivariate receptor models.

Watson (1979) has carried through the calculations of the source-receptor model relationship for the correlation and principal components models in forty-three equation-laden pages. Only a few highlights can be offered here.

The mathematical obfuscation of these models must not remove the requirement that every receptor model must be representative of and derivable from physical reality as represented by the source model. A statistical relationship between the variability of one observable and another is insufficient to define cause and effect unless this physical significance can be established.

The multivariate models deal with a series of m measurements of aerosol component i during sampling period or at sample site k. From Equation 3,

$$C_{ik} = \sum_{j=1}^{p} a_{ij} S_{jk} \qquad k=1...m \tag{12}$$

The multivariate models deal only with the C_{ik} with the objective of predicting the number of sources, p, and which a_{ij} are associated with which S_j, or more ambitiously, estimating a_{ij} and S_{jk}.

The linear regression model sets

$$\hat{C}_{vk} = a\, C_{uk} + b \tag{13}$$

where

$$a = \frac{\overline{C_v C_u} - \overline{C}_v \overline{C}_u}{\overline{C_u^2} - \overline{C}_u^2} \tag{14}$$

and

$$b = \overline{C_v} - a\,\overline{C_u} \tag{15}$$

with

$$\overline{C_v C_u} = \frac{1}{m} \sum_{k=1}^{m} C_{vk} C_{uk} \tag{16}$$

$$\overline{C_u} = \frac{1}{m} \sum_{k=1}^{m} C_{uk} \tag{17}$$

$$\overline{C_u^2} = \frac{1}{m} \sum_{k=1}^{m} C_{uk}^2 \tag{18}$$

and $\hat{C}_{vk}$ is the value calculated from Equation 12 as opposed to the measured value, C_{vk}.

When chemical components u and v originate uniquely in source type j, C_v and C_u can be replaced by Equations 4 in Equations 14 and 15 to give

$$a = \frac{a_{vj}}{a_{uj}} \tag{19}$$

$$b = 0 \tag{20}$$

The linear regression model yields the ratios of the source compositions for chemical components unique to one source.

The correlation model relies on the correlation coefficient,

$$r_{C_u C_v} = \frac{\overline{C_u C_v} - \overline{C_u}\,\overline{C_v}}{\left(\overline{C_u^2} - \overline{C_u}^2\right)^{1/2}\left(\overline{C_v^2} - \overline{C_v}^2\right)^{1/2}} \quad (21)$$

to measure the extent to which ambient concentrations u and v vary in the same way. If u and v are from the same source, then Equation 4 holds and upon incorporation into Equation 21

$$r_{C_u C_v} = 1 \quad (22)$$

The traditional factor analysis model transforms equation 12 to

$$C_{ik}^s = \sum_{j=1}^{p} \sigma_{ij}\, S_{jk}^s + d_j\, U_{ij} \quad (23)$$

where

$$C_{ik}^s = \frac{C_{ik} - \overline{C}_i}{\left(\overline{C_i^2} - \overline{C}_i^2\right)^{1/2}} \quad (24)$$

$$S_{jk}^s = \frac{S_{jk} - \overline{S}_j}{\left(\overline{S_j^2} - \overline{S}_j^2\right)^{1/2}} \quad (25)$$

and

$$\alpha_{ij} = a_{ij}\left(\frac{\overline{S_j^2} - \overline{S}_j^2}{\overline{C_i^2} - \overline{C}_i^2}\right)^{\frac{1}{2}} \quad (26)$$

where α_{ij} is called the factor loading, and is related to the source compositions through Equation 25, and S_{jk}^s is called the factor score, which is related to the source contributions through Equation 24. The d_j and U_j are unique factor loadings and scores, respectively, which are often left out of the analysis. The resulting is a principal components analysis

$$C^s_{ik} = \sum_{j=1}^{p} a_{ij} S^s_{jk} \qquad (27)$$

The factor model treats the C^s_{ik} as components of a vector for each chemical component, i, in an m-dimensional space. If Equation 3 is true, then only p vectors, where $p \leq m$, in a less complex p-dimensional space are required to produce the vectors of C^s_i. This p-space is defined by the eigenvectors of the correlation matrix of the C^s_i. These p eigenvectors merely define the space, however, and are not necessarily representative of the sources, the S^s_j vectors. They must be linearly combined to form the new source vectors.

Alpert and Hopke (1980) have experimented with an unstandardized correlation matrix with correlations between samples as opposed to the standardized correlation between chemical species of Equation 21. This approach retains more of the information in the data and allows the proportionality among the chemical species to be maintained.

The foregoing discussion can by no means initiate the casual reader to the full power and limitations of the multivariate models; it can only impress on him the complexity of the subject and the necessity to study it carefully before proceeding.

Multivariate models have been successful in identifying source contributions in urban areas. They are not independent of source composition knowledge since the chemical component associations they reveal must be verified by source emissions analyses. The linear regression model can produce the typical ratio of chemical components in a source, but only under fairly restrictive conditions. The factor and principal components analysis models require source composition estimates to find the right set of S^s_j vectors, though it seems that these a_{ij} estimates can be refined as a result of the analysis (Henry, 1977, Alpert, 1980).

The most important caveat to be heeded in the use of multivariate models is that though origination in the same source will cause two chemical components to correlate, the converse, that chemical

components which correlate must have originated in the same source, is not true. The substitution of Equation 1 into Equation 13 and its propagation through the subsequent equations shows that the correlation between elements u and v is a function of the non-source related dispersion factor, D_{ik}, as well as the source-dependent emissions factor, E_{ik}.

The only input data required of the multivariate receptor models at their present state-of-the-art are the ambient concentrations, the C_{ik}. This data set must be large (just how large is yet undefined) and must contain substantial variability. With an estimate of the a_{ij}, the source compositions and contributions can be refined. No provision has yet been made for propagating the measurement uncertainties, the σ_{C_i} and $\sigma_{a_{ij}}$, through the model or of evaluating their effect on model output as has been done for the chemical mass balance.

Future development of the multivariate models includes:

- Testing of other multivariate methods. Suggestions include ridge regression (McDonald and Schwing, 1973) regression against principal components (Henry and Hidy, 1979), clustering techniques (Gaarenstroom et al., 1977), time series analysis (Box and Jenkins, 1976), and non-parametric (Bradley, 1968) approaches. The formulation and testing of such models should draw statisticians and applied mathematicians into working with aerosol study specialists to become aware of the full range of multivariate methods available and to apply them appropriately to air quality problems.

- Creation of simulated ambient data sets from a simple source model with known source compositions and contributions perturbed by typical experimental error. The multivariate models should be applied to these data sets, as did Watson (1979) for the chemical mass balance, to determine the extent to which the models can apportion source contributions under various conditions.

2.3 Microscopical Identification Models

Microscopic identification and classification of individual aerosol particles into source categories can be a valuable tool for the study of settled and suspended atmospheric particles.

Many different optical and chemical properties of single aerosol particles can be measured, enough to distinguish those originating in one source type from those originating in another. The microscopic analysis receptor model takes the form of the chemical mass balance equations presented in Equation 3.

$$M_i = \sum_{j=1}^{p} a_{ij} S_j \qquad (28)$$

where

M_i = % mass of particle i in the receptor sample
a_{ij} = fractional mass of particle type i in source type j
S_j = % of total mass measured at the receptor due to source j.

The model of Equations 3 and 28 might be better termed an aerosol properties mass balance to encompass both the chemical mass balance and microscopic receptor models.

The solutions to the set of Equations 28 are the same as those described by Equations 4, 5 to 7, 8, and 9, though only the tracer method, i.e., each receptor particle type is unique to one source type, has been used in the past.

The microscopic receptor model can include many more aerosol property measurements than those used to date in the chemical mass balance and multivariate models. It has not taken advantage of the mathematical framework developed in the other two models to deal with particle types which are not unique to a given source type or the ambient and source measurement uncertainties.

The data inputs required for this model are the ambient properties measurements, the M_i, and the source properties measurements, the a_{ij}. To estimate the confidence interval of the calculated S_j, the uncertainties σ_{M_i} and $\sigma_{a_{ij}}$ are also required. Microscopists generally agree that a list of likely source

contributors, their location with respect to the receptor, and windflow during sampling are helpful in confirming their source assignments.

The major limitation of microscopic receptor models is that the analytical method, the classification of particles possessing a defined set of properties, has not been separated from the source apportionment of those particles. Equations 28 have never been used in this application. The source identification takes place on recognition of the particle by the microscopist. The particle properties he uses for this identification are his alone (or his laboratory's) and are not subject to interpretation by another.

Source contributions assigned to the same aerosol sample have varied greatly in inter-comparison studies (Bradway and Record, 1976), but without the intermediate particle property classifications, it is impossible to ascribe the differences to the analytical portion or to the source assignment portion of the process.

Many times the microscopist relies on his past knowledge of the properties of aerosol sources without the examination of local source material. For example, a coarse particle sample taken in the vicinity of a coal fired power plant may show a 20% contribution due to flyash and an 80% contribution due to minerals. As Brookman and Yocom (1980) point out, the 20% flyash may not have come from the power plant during the sampling period. An examination of nearby soils could show a 20% flyash concentration resulting from long-term deposition. Using one of the least squares or linear programming approaches to the solution of a set of Equations 28 with local source compositions could alleviate this problem.

Though the aerosol properties mass balance suggested by the microscopic models shows promise, it is limited by the lack of a standardized, reproducible analytical method. A few laboratories are pursuing the establishment of such a method, however, and the state-of-the-art of their efforts will be summarized in Chapter Three.

Future development for this model include:

- Creation and acceptance of a standardized methodology.
- The definition of important optical criteria and their measurement/methods.
- A method of reporting and cataloging source properties by means of the chosen criteria.

2.4 Hybrid Source/Receptor Models

Until now, the receptor models have been treated as if they were completely separate entities from the source models. This need not be the case. Three examples of source/receptor model hybridization show promise, and many others are probably waiting to be developed. A source model incorporates measured or estimated values for the emission rate, E_j, and the dispersion factor, D_j, in Equation 1. Whenever either of these enter the receptor model as observables, it shall be termed a hybrid model. The three applications considered here are emission inventory scaling, micro-inventories, and dispersion modeling of specific sources within a source type.

Emission inventory scaling, proposed by Gartrell and Friedlander (1975), uses the relative emission rates of two source types subject to approximately the same dispersion factor (e.g., residential heating by woodstoves and natural gas) to approximate the source contribution from the source type not included in the chemical mass balance (e.g., natural gas combustion). The ratio of the emission rates is multiplied by the contribution of the source type which was included in the balance.

The micro-inventory approach developed by Pace (1979) has been shown to be a good predictor of annual average TSP based on a detailed assessment of emissions close to the sampling site. Four types of emissions are compiled using standard emission factors where applicable.

- o AREA emissions: annual area source particulate matter emissions within one mile of the monitor. This variable includes fugitive dust and the influence of the monitor height.

- POINT emissions: annual point source emissions within five miles of the receptor.
- LOCAL emissions: annual emissions from traffic-related sources within 200 feet of the receptor.
- VISPLUME emissions: an indicator variable for the presence or absence of traffic related/suspended dust plumes from nearby (200 ft.) streets.

Pace investigated many other variables in multiple linear regressions with TSP, but found these to be the significant ones.

Figure 2 shows the excellent relationship between predicted and measured values for the 79 data sets from four cities used by Pace to derive the following relationship:

$$\text{Annual Average TSP} = 0.0045\,(\text{AREA}) + 0.00096\,(\text{POINT}) + 50.5\,(\text{LOCAL}) + 18.6\,(\text{VISPLUME}) + \text{CITY EFFECTS} \quad (29)$$

Pace emphasizes that Equation 29 is not applicable to other cities in an absolute sense, but he does show, with data from three other cities, that it is effective in determining the relative contributions of AREA, POINT, LOCAL, and VISPLUME effects. Large contributions from LOCAL and VISPLUME indicate that the sampler location is probably inadequate to represent the airshed as a whole, and that the best control measures are those taken in the vicinity of the monitor.

A more specific equation than Equation 29 could be calculated by adding new data sets to the 79 already in use and recalculating the coefficients, or by using the existing HIVOL sites to create a new equation.

The micro-inventory should be a pre-requisite for aerosol mass balance or microscopic model applications to identify the most likely sources for sampling, analysis and inclusion in the balance.

A major limitation of receptor models is their inability to distinguish between specific sources within a source type. Resuspended road dust may be a major cause of standard violations, but until the offending roadways can be pinpointed, a control strategy

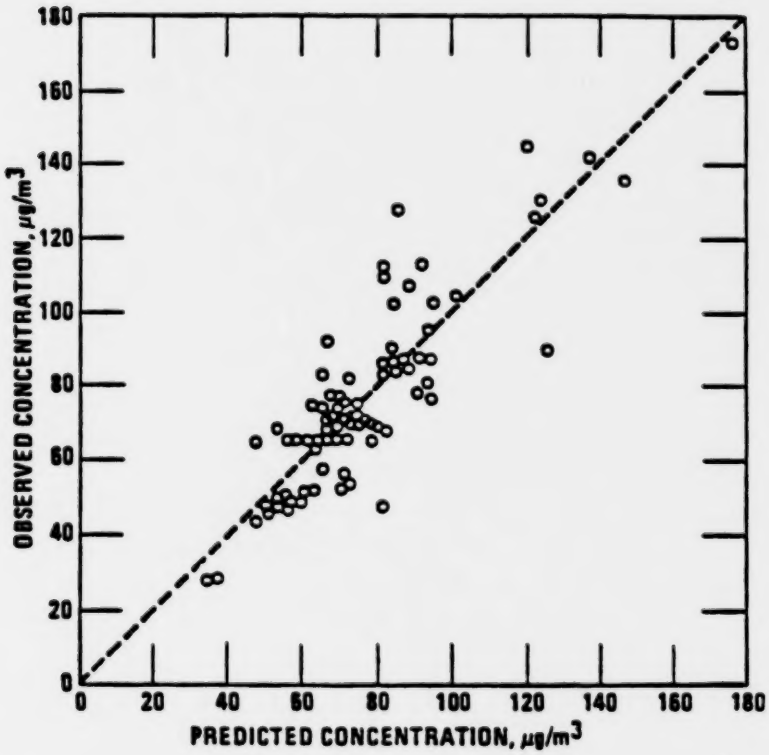

Figure 2. Plot of predicted versus observed concentration for regression. From Pace (1979)

cannot be implemented. A major limitation of source models is the necessity to estimate emission rates from the many, diverse producers of suspended particulate matter. Figure 3 schematically illustrates a possible combination of the two models. The receptor model quantifies the source type contributions. Only the major contributors need to be evaluated for the source model, so that resources which might have been used to inventory an entire airshed can be concentrated on determining emission rates from the specific emitters within a source type. This improved data with fewer source inputs should increase the accuracy with which the relative contribution from each individual source type can be calculated. An application of this hybridization has never been attempted, but it merits exploration.

Two more sets of observables are introduced into the hybrid models, the emissions factors, the E_j, and the dispersion factors, the D_j. It is the difficulty of quantifying these that led to the use of a receptor model over the source model in the first place, so it would seem there is little advantage in re-introducing them. The advantage of the hybridization is that the number of E_j and D_j can be considerably reduced and that the relative values rather than the absolute values are used. These relative values are more accurate in most cases.

Still the uncertainties σ_{E_j} and σ_{D_j} need to be evaluated and incorporated into any source/receptor hybrid model.

Future areas of development include:

- Theoretical formulation of source/receptor models.
- Validation tests with real and simulated data.

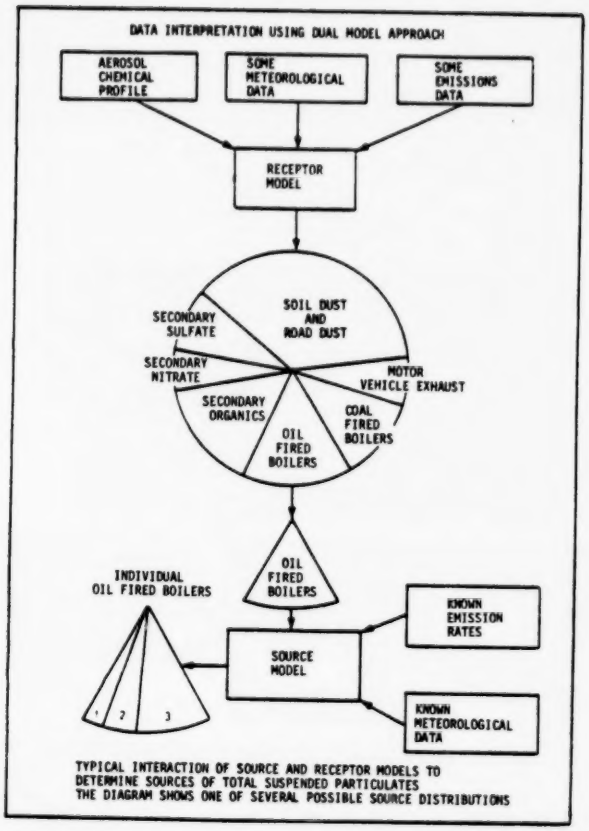

Figure 3. This source receptor model hybridization uses the receptor model to calculate the contributions of each source type and the source model to quantify the contribution of each specific source to its source type. (From ERT, 1979)

CHAPTER 3. RECEPTOR MODEL INPUT DATA

It is evident from the previous chapter that the receptor models, like all such simplified representations of reality, can return results no better than the input data with which they are supplied.

The measurements required for the present receptor model state-of-the-art include particulate matter composition, size and variability of both source and receptor. Aerodynamic diameter and variability must be distinguished at the field sampling stage (though microscopic sizing can provide an *a posteriori* estimate of optical diameters). While real time measurements of particle composition would be ideal, the state-of-the-art requires laboratory analysis of particulate matter collected on appropriate substrates.

In this chapter, the collection of input data for receptor models will be examined.

3.1 Field Study Design and Data Management

Few aerosol characterization studies performed in the past have been designed with the goal of applying a receptor model. The receptor models have traditionally been developed and applied *after* data collection as a method of interpreting large, complicated sets of measurements. This has been a productive development mechanism, but the present state-of-the-art demands a more precise definition of the goals of a study, the receptor models to be used, the input data required, and the measurement and quality assurance program to obtain that input data. These goals might include:

- studies to further validate the receptor models and measurement methods,
- evaluation of health related particulate matter constituents,
- causes of visibility impairment, and
- providing a detailed breakdown of source contributions to different particle size fractions in order to devise a control strategy.

Cost will be the primary controlling factor in the experimental design in essentially all cases. The cost of obtaining the entire population of possible measurements is prohibitive. The field study design must select a subset which is representative of the population within definable confidence limits and which can be obtained within the constraints of existing resources.

Several generalizations can be made about aerosol characterization study design based on past experience.

Existing data should be used to obtain an understanding of the area under study. These data include historical HIVOL and gas concentrations, meteorological data, emission inventories, chemical and optical analyses, and the results of dispersion modeling. Step-by-step procedures of how to use this information need to be developed.

Siting of monitoring stations should take advantage of existing sites with consideration to land use categories and transportation source influences. Prevailing wind directions and the locations of important sources should be located prior to siting and used in the siting process. Source-oriented sites are often useful, but both upwind and downwind, baseline or background sites are needed. Simple source-oriented modeling can provide helpful guidance for the location of sites. Hougland and Stephens (1976) describe an analytical technique for siting monitors while Stalker and Dickerson (1962) and Solomon et al. (1977) offer empirical evidence that a small network can represent an airshed as well as a larger one under certain conditions. In many cases of current interest, sampling for vertical stratification may be needed to distinguish between tall stack and ground level contributions.

The frequency and duration of measurement are important factors; source contributions at a fixed station are often short-term and to measure their variability requires sampling times of the same order as that variation. Twenty-four hour sampling is considered minimal, and shorter term sampling is often desirable. Less than 12-hr samples are usually required to relate samples to a constant wind direction.

Measurements are needed to extrapolate to annual average conditions; however, it is not necessary to sample daily to achieve this. It is important to obtain information on both intense pollution conditions (episodes) and relatively clean conditions. Thus, more samples than needed should be taken with a subset chosen for detailed analysis based on meteorological or other factors. Samples should be taken every day over selected seasonal periods rather than one per third or sixth day to capture the progression effect of multiday events.

Aerosol sampling devices must be chosen which can obtain an adequate amount of particulate matter for analysis within the sampling duration. The samplers must separate the ambient aerosol concentrations into the desired size ranges with a defined efficiency.

The particle size cuts of the sampler and their variation with respect to windspeed and wind direction (McFarland et al., 1979) must be quantified if comparability of results between one study and another is to be achieved. Several of the sampling devices available for aerosol characterization studies were intercompared via the simultaneous sampling of air during sixteen consecutive twelve-hour periods (Camp et al., 1978). More of this testing and intercomparison work is required of existing samplers.

For sampling in which the variation of aerosol concentrations with time is required, the sampler should be able to switch filters sequentially without operator attention. Camp et al. (1978) report three commercially available samplers, the automated dichotomous sampler, the sequential filter sampler, and the streaker sampler, capable of this.

Because of the re-entrainment and bounce-off problems associated with cascade impactors, which cause large particles to penetrate to the small particle stages, virtual impactors (Loo et al., 1975) and cyclone preseparators (Mueller et al., 1980) are the most commonly used size separating devices in current aerosol characterization studies.

In most studies, particularly those in which regulatory issues are addressed, a 24-hour high volume sample should accompany samples taken with other devices; the relationship of the aerosol study samples to the historical total suspended particulate matter data base can then be evaluated.

Meteorological information which has been useful to receptor model studies includes temperature, relative humidity, mixing height, windspeed and wind direction. For hybrid approaches using a source model, stability class, sigma theta and vertical temperature profiles may be needed. The wind direction is particularly important for the verification of certain receptor model source contribution predictions; definite differences should exist between samples on which the source is upwind and downwind of the receptor. Kleinman (1977) incorporated a normalization factor, which is a function of windspeed and mixing depth, to minimize dispersion related correlations from his multivariate receptor model. Existing monitoring at airports, television stations and private facilities may be sufficient for the aerosol study. These networks, their ability to meet the requirements of the study, and the data collection procedures need to be defined as part of the field study design.

Source characterization must be an integral part of any field study design. A microinventory (Pace, 1979) of each sampling site should be conducted and specimens of nearby fugitive emissions sources identified by it should be taken and analyzed. Aerosol properties of likely point source contributors identified in the emission inventory must be measured during the course of ambient sampling. Source sampling and analysis techniques need to be compatible with ambient techniques. With limited resources, not all sources can be characterized.

Priority should be given to those sources which are expected to be heavy contributors on the basis of calculated emissions (EPA, 1973), and which have not been characterized in previous studies.

The chemical components to be quantified in each sample and the analytical methods to be used should be specified before the first sample is taken. The experimental design must be reviewed to assure that the field samples will be amenable to the chosen techniques. This requires:

- Comparison of component concentrations likely to be found in samples to the lower quantifiable limits of the analytical methods. The sampling design or analytical method must be modified if the majority of the expected amounts are less than these limits.

- Selection of filter media which will not bias the aerosol measurement because of variable blank concentrations or artifact formation of the chemical species to be measured. If x-ray fluorescence is to be used, the aerosol must be deposited on the surface rather than inside of the filter to prevent absorption of x-rays by the filter material. Simultaneous sampling on more than one substrate may be required.

- Transport and storage procedures must prevent changes in the components to be quantified between sampling and analysis.

An overwhelming number of chemical compounds can be found in the typical urban aerosol, and some subset must be selected for quantification. Two general rules can narrow the scope of chemical analysis somewhat. First, the sum of the masses of chemical components measured should equal, within stated precision estimates, the total mass concentration of the aerosol. Without this equivalence one cannot necessarily assign the unaccounted for mass to a source, nor is it possible to verify the other component quantities via a mass balance. Thus the first requirement of chemical analysis is that the major chemical components of total aerosol mass in the sample are quantified.

The second requirement is that components unique to specific aerosol sources affecting the ambient sample are measured. The precise chemical species will depend on the sources in the airshed under consideration.

Most studies will require measurement of certain "basic" particulate matter species including ions, elements, and organic and inorganic carbon.

A quality assurance component must be integral to the study to verify the accuracy of its measurements and to estimate a precision for each one. This component should include co-located sampling, replicate analysis, station audits, data validation, interlaboratory comparisons and a full set of standard operating procedures. The quality of the data set generated should be discussed and should include the results of validation, analysis of outliers and overall estimates of accuracy and precision.

Every data item should be considered an interval rather than a number and the precision of each measurement determined from replicate analysis should be propagated through all calculations (Bevington, 1969).

The final important ingredient of experimental design is data management, which is normally carried out by computer. The data must be organized and merged into uniform formats accessible by the receptor models. Provisions for the entry and processing of quality assurance data will greatly reduce the efforts required for this often neglected study component. All data from initial analyses must be retained until the sorting of relevant and irrelevant information can be achieved. Economical accessibility, security and definition of uncertainty should be keynotes of data management systems for this work. Organization techniques for "data dipping" subsets of data from a number of large data bases to a microcomputer must be developed to maximize economic data handling and computation.

Though the field study design task is presented here as a separate entity, it requires a knowledge of all other aerosol study aspects and dictates which subset of the array of tools available will accomplish the goals of the study within the resources available. No two field study designs will be alike, but studies with similar goals should be similar in design.

Several generalized "ideal" or "template" designs to meet the needs of users with a specific set of goals would be helpful examples to those designing a field study for the first time.

Future development efforts in field study design and data management should include:

- Develop methodologies for choosing sampler location, sampling schedule, and sampling devices.
- Measure the collection efficiency of available samplers as a function of particle size, windspeed and wind direction.
- Create a series of "typical" design scenarios which can be specified for different aerosol study objectives.
- Develop standardized data bases from which ambient and source information can be "data dipped."

- Develop standarized receptor models for computer implementation and standard interfaces to the relevant data bases.

- Create quality assurance procedures and methods of reducing the data to assign a confidence interval to each measured quantity.

3.2 Source Characterization

All receptor models, even the source/receptor hybrids, require input data about the particulate matter sources. The multivariate models, which can conceivably be used to better estimate source compositions, require an initial knowledge of the chemical species associations in sources. The lack of knowledge of a_{ij} and σa_{ij} in Equations 3, 8, and 9 poses the greatest limitation to receptor modeling.

Existing data on characteristics of particles from various types of sources are inadequate for general use, though they have been used in specific studies with some success. Most of the source tests have been made for purposes other than receptor modeling and complete chemical and microscopical analyses have not been completed. Source operating parameters which might affect the aerosol properties of emissions have not been identified nor measured. Particle size separations are rarely the same as those measured in ambient sampling and no provision is made for likely transformations of the source material when it comes into equilibrium under ambient conditions.

For receptor modeling, the only source characteristics that are relevant are those perceived at the receptor. But if source components could be separated from each other at the receptor, there would be no need for a receptor model.

Stack sampling is not sufficient for characterizing all sources and may not be the best method even for ducted point sources. Because of condensation of vapors, fallout of very large particles and the neglect of non-ducted emissions, the materials collected from stacks, especially those at high temperatures, will often not represent the particles from the source as observed at a receptor several kilometers away.

Several source type categories, such as road dust, automobile exhaust and home heating, contain many individual sources whose emissions compositions may vary considerably. A source sampling program for these source types must consider the selection of a representative subset as well as the sampling mechanics.

At present, most source sampling groups measure elemental, organic or microscopic components, each depending on its area of expertise. No complete set of measurements on a single sample exist.

In spite of this criticism of existing source characterization data, knowledge of several source types and testing methods is growing. Certain aerosol property measurements have been made on emissions from geological material (Rahn, 1976), auto exhaust (Pierson and Brachaczek, 1974), marine background (Hidy et al., 1974), coal combustion (Gladney et al., 1976), municipal incinerators (Greenberg, 1978), iron and steel blast furnaces (Jacko, 1977), tire dust (Pierson and Brachaczek, 1974), copper smelters (Zoller et al., 1978), vegetative burning (Oregon Department of Environmental Quality, 1979), kraft recovery boilers (Watson, 1979) and cement plants which, though semi-quantitative for the most part, have been useful in applying receptor models. These source characterization experiences point out some of the considerations which are required of receptor model source sampling.

For use with receptor models, it is desirable to collect particles far enough from the source to allow the emissions to reach equilibrium with the atmosphere, but not so far away that the source material under test becomes contaminated with aerosol from other sources.

Armstrong et al. (1978) describe a unique approach of attaching sampling equipment to tethered balloons which are maneuvered from the ground into the plume of smokestack emissions. The payload these balloons can carry is presently low, but conceivably it could be increased to allow the placement of size-selective sampling devices in the plumes of tall point sources.

Aircraft sampling in plumes (Mroz, 1976) has long been used, but it is expensive and difficult. Sampling flowrates need to be increased because of the short period of time an airplane can remain

in the plume. A real time measurement device, such as a nephelometor, condensation nuclei counter or SO_2 monitor, is required to inform the sampler when it enters and leaves the plume so that sampling can be started and stopped accordingly. Flowrates must be changed with airspeed to maintain isokinetic conditions - this makes size-selective sampling difficult. The high cost and trouble of aircraft sampling do not encourage its widespread use for receptor model source characterization.

A promising development is that reported by Rheingrover and Gordon (1980). Using ambient samplers located in "maximum effect" areas for specific sources and wind flow patterns, they have distinguished the composition of a single point source at a receptor. They then use these source characteristics to represent that source in areas less affected by that source, where its contribution is not so obvious. This approach could be incorporated into field study designs in the following way:

1. After identifying major point sources, determine the "maximum effect" areas via source modeling.

2. Locate samplers and windvanes at these sites and sample simultaneously with the other receptor sites.

3. Analyze samples for all days on which the sampler is downwind of the source.

4. Determine which components of the sample are due to the source in question.

The last point is the hardest to accomplish and reverts to the question of, "If that can be done, who needs the receptor model?" The true advantage of having a sampler in the "maximum effect" area may be in eliminating the particular source it is intended to monitor from consideration in the receptor modeling at other sites; if it cannot be detected where it should hit hardest, then there is little hope of finding it elsewhere.

In-stack testing procedures are still under development with a greater inclination on the part of developers to produce samples of use for receptor modeling. In the past, few stack samples have

obtained more than mass emission rates under stack conditions. New sampling methods include dilution with clean air to simulate ambient conditions, filter media amenable to chemical analyses, and samples in size ranges similar to those sampled in ambient air (Bruce Harris, EPA/IERL, personal communication, 1980).

Detailed studies of large point sources should be done in such a way as to enhance the generality of the results. The relevant operating parameters affecting emission compositions need to be identified, measured and reported with each source test. It is only with this information that the test results can be evaluated for use in receptor model studies other than that for which the test was made.

Studies of motor vehicle emissions are difficult now and will become even more so as lead is phased out of gasoline. Studies of tailpipe emissions should be done with dilution and cooling of the exhaust. A serious drawback of individual automobile testing is the variability of emissions compositon as a function of car and driving cycle. In his review of the automobile emissions literature, Watson (1979) found the lead composition of auto exhaust varying from 12 to 70 percent, depending on the test.

Studies in tunnels (Pierson and Bracheczek, 1976, Ondov et al., 1979) and along highways (Feeney, et al., 1975), might offer more representative compositions of the motor vehicle fleet affecting receptors. At present, no components unique to diesel exhaust have been found, and with the phase-out of lead in gasoline, the major tracer for conventional engines is being lost. Much greater emphasis on the characterization of this universal aerosol source category is required.

The characterization of fugitive emissions, those originating from roadways, storage piles, and rock crushers, is extremely important for the source apportionment of the total and coarse suspended particulate matter fractions of ambient concentrations. Once likely sources are identified, ambient samples can be taken at site or bulk material can be taken to the laboratory, resuspended and sampled on a filter substrate and analyzed. Analysis of the bulk material without resuspension may yield a biased characterization

since the larger particles of the bulk sample which do not remain resuspended often have compositions differing from the smaller particles which do remain suspended (Rahn, 1976).

Source characterization results are not located in a centralized facility which is constantly updated, and each researcher has to assemble his own collection of results or use the assemblage of someone else which may not be applicable to the case under study. Fortunately, the Environmental Protection Agency has established the Environmental Assessment Data System (EADS) (Larkin and Johnson, 1979) which contains chemical compositions of particulate matter emissions tests. This existing computerized structure can provide the centralized location for receptor model source characterization information. Procedures such as those described for ambient data in the previous section, need to be developed which will allow receptor model users to access this data base over telephone lines. The data required of receptor model source tests should be incorporated into the EADS and source characterization results should report this information in an EADS compatible format.

Future development efforts in source characterization should include:

- Develop new source sampling methods including tethered balloon and ground based sampling.
- Standardize data reporting and management procedures and store all data in a central data base.
- Create a chemical component analysis protocol to obtain maximum information from each source test.

3.3 Analytical Methods

Many chemical and physical analysis methods exist to characterize particulate matter collected on a substrate. Though several methods are multi-species, able to quantify a number of chemical components simultaneously, no single method is sufficient to both quantify the majority of the collected particulate matter mass and those components which serve to identify and quantify source contributions.

After the composition measurements to be made have been selected, each analytical method must be judged in terms of four necessary and three desirable criteria. These apply not only to the analytical method, but to the laboratory performing the method.

The necessary criteria are:

1. The analytical method must have been developed and tested specifically for the analysis of suspended particulate matter.

2. Lower quantifiable concentrations of the method must be less than the concentrations expected from the ambient samples taken.

3. The method must be free of biases for all components quantified.

4. The values achieved by the method must be reproducible within defined and reasonable confidence intervals.

If an analytical method is successful for certain types of environmental samples, it is convenient to assume that it is appropriate for all samples. Hard and bitter experience shows this not to be the case for suspended particulate matter. Before a procedure can be accepted, one must have identified and corrected likely interferences, defined amenable aerosol collection substrates and sampling requirements, considered the effects of different aerosol matrices, and verified its ability to meet the final three criteria.

The amount of suspended particulate matter available for analysis depends on the ambient concentration, the flowrate of air through the substrate, the sampling duration, and the fraction of the sample presented for analysis. Cooper (1973) provides a useful tabulation of typical concentrations and ranges of urban aerosol elemental concentrations; it would be helpful to update this summary, expand it to certain chemical compounds, and stratify it by site types (urban, rural, etc.). The expected amounts available for analysis should be a factor of ten or more above the lower quantifiable concentrations of the analytical method.

Measurement accuracy can only be established through interlaboratory/intermethod comparisons and the analysis of standard reference materials. Intermethod comparisons are better for evaluating accuracy since interlaboratory analyses using the same method will exhibit the same types of biases. Urban Particulate Matter Standard Reference Material 1648 (Greenberg, 1979) provides a means for initial and routine verification of accuracy. The concentrations of additional chemical components in this standard need to be quantified and their constancy with time assessed. NBS 1648 was collected in St. Louis and is representative of the eastern city aerosol matrix. Other aerosol standards from a variety of locations would be helpful for the evaluation of analytical bias.

Replicate analyses of the same sample must return results with a standard deviation of $\pm$ 10% or less at concentrations greater than ten times the lower quantifiable limit. The data from replicate analysis should be used to define the precision of each value measured.

The desirable criteria are:

1. The method should measure a number of components in a single analysis.
2. The analysis should be non-destructive.
3. The analysis should be cost-effective.

Many analysis techniques are capable of measuring more than one chemical component of interest with adequate precision, accuracy and sensitivity. Not only do these methods save the cost of many individual analyses, they minimize the amount of sample required.

A non-destructive method is preferred since the sample can be made available for other analyses to verify the non-destructive results or to complement them with the measurement of additional chemical components.

Given the choice of analytical methods meeting the necessary and desirable criteria, the least expensive analysis per sample should be chosen. This choice may vary with the number of samples to be processed.

The range of analytical methods available for particulate matter analysis is large; many of the available methods are critically reviewed by Katz (1980). A complete review of all methods applied in the past with a view toward their use for receptor modeling does not exist, and its production would be a useful contribution to the state-of-the-art. Several analytical tools have been found to meet the stated criteria, or are in the development stages of trying to meet them, and are being used to supply the input measurements for receptor models. One means of classifying available and useful analysis procedures for source and receptor studies consists of the following categories:

1. Elements with atomic number greater than 11.
2. Carbon
3. Ions
4. Organic Compounds
5. Inorganic Compounds
6. Physical and Optical Properties

Several specific methods belonging to each category and the extent to which they meet the stated criteria are discussed here because of their widespread acceptance and use in the field of receptor modeling. Analytical methods not mentioned here should not be dismissed; they are also potentially useful. As the extent to which they meet the necessary and desirable criteria becomes known, their use for obtaining receptor model input data will increase accordingly.

Elements

X-ray emission spectroscopy (Giaque et al., 1974, Cahill et al., 1976) (photon or proton excited) has developed as a reliable, available, cost-effective and non-destructive elemental analysis method. More aerosol samples, numbering in the hundreds of thousands, have been analyzed by this method than by any other. The method

requires aerosol samples uniformly deposited on a thin, flat organic substrate (Teflon, nuclepore, polycarbonate and cellulose acetate membrane filters are commonly used). In typical urban samples from 10 to 20 elements are routinely measured. Biases exist due to the attenuation of light element (e.g., Al and Si) x-rays in coarse (>2 μm) particles, but corrections (Dzubay and Nelson, 1974) have been devised and applied. HIVOL samples on fibrous filters are inappropriate for this method because a portion of the aerosol penetrates beyond the filter surface and is collected within the volume of the filter material; the x-rays emitted by these particles are partially absorbed by the filter material. Spectral interferences of one element x-ray with another must be accounted for in the data reduction, but this is usually handled by the computer based spectrum processing routine (Bonner et al, 1973). Interlaboratory and intermethod comparisons (Camp et al., 1974, 1975) verify its accuracy and precision.

Instrumental neutron activation analysis (Zoller et al., 1970, Dams et al., 1970) is also reliable, available, cost-effective and non-destructive (though substantial decay times may be required before the sample can be submitted to other analyses). It equals or surpasses x-ray emission in sensitivity for many elements and allows the quantification of a number of rare-earth elements not detectable by other methods. It is more labor intensive than x-ray analysis and therefore more costly. The aerosol deposit need not be confined to the substrate surface since high energy gamma rays experience no absorption in the filter material. Suspended particulate matter on glass fiber substrates have been analyzed by neutron activation (Lambert and Wilshire, 1979) but due to the high and variable blank concentrations of certain elements in these filters their use is not recommended (Gilbert and Fornes, 1980). Several fibrous filters appropriate for HIVOL sampling (Dams et al., 1972) have been evaluated and found satisfactory. Most spectral interferences can be eliminated by optimizing irradiation time, decay time and counting time. Camp et al. (1974, 1975, 1978) have verified the accuracy and precision of the method in interlaboratory comparisons.

Atomic Absorption (AAS), Atomic Emission, (AES) and Inductively Coupled Plasma/Atomic Emission Spectroscopy (ICP/AES) (Ranweiler and Moyers, 1972, Olson et al., 1978, Lynch et al., 1980) are destructive methods, requiring sample dissolution, with the requisite sensitivity for suspended particulate matter analysis. The AAS and AES analyses are single element methods while ICP/AES is multi-element. The method is applicable to aerosol collected on fibrous as well as membrane filters, though glass fiber substrates with large and variable blank concentrations bias analytical results. The particulate matter must be totally dissolved with a minimum loss of elements through volatilization; more than one extraction procedure may be required, depending on the elements sought. McQuaker et al. (1979) and Ranweiler and Moyers (1972) describe extraction procedures and their successful applications to standard reference materials. The variable dissolution of particulate matter might be used to identify the compounds in which elements occur through a series of sequential extractions (Tessier et al., 1979); this approach has not been tried for suspended particulate matter. Intermethod comparison results of particulate matter samples are available for only a few elements (Camp et al., 1975, Walling et al., 1978, Barbaray et al., 1979); the results of these intercomparisons are variable, with some elemental concentrations being reported factors of five and six lower than the values reported by the other participating laboratories. A systematic intecomparison with the standard urban aerosol for a variety of extraction procedures is needed.

Other elemental analysis methods, including spark source mass spectrometry are available and their appropriateness for suspended particulate matter analysis needs to be evaluated.

Carbon

Though carbon is indeed an element, its quantification is not amenable to the aforementioned methods. Because of its large contribution to both source and ambient suspended particulate matter, its concentration in samples is important. Combustion in oxygen followed by CO_2 measurements, or conversion of CO_2 to CH_4 and

measurement with a flame ionization detector, can provide reproducible and quantitative total carbon values. Different atmospheres and combustion temperatures can provide an operational separation of elemental and organic carbon (Johnson and Huntzicker, 1978). Nuclear (Macias et al., 1978), solvent extractions (Appel et al., 1976, 1979), and light absorption methods have been devised. Macias et al. (1978) report some intermethod comparison results, but more interlaboratory work is needed. Carbon values for the NBS 1648 are not reported and accepted values need to be obtained.

An important development in the measurement and classification of carbon for receptor models is the quantification of the radioactive C^{14} content of suspended particulate matter by low-level counting, (Currie et al., 1978). Carbon 14 is a unique tracer for carbon from recently living material; it is nearly non-existent in emissions from fossil fuel sources.

Carbon analyses should be performed on non-carbon containing substrates, though there are possibilities of subtracting the filter blanks (R.K. Stevens, USEPA, personal communication, 1980).

Ions

A wide variety of ion analysis methods exist with adequate lower quantifiable concentrations. All are destructive, requiring dissolution of a portion of the sample. For receptor model studies, the multi-element capabilities of ion chromatography (Small, 1975) and automated colorimetry (Technicon, 1972) have resulted in the greatest volume of suspended particulate matter ion analyses being performed by one of the two methods. Practically all collection substrates are amenable to these analyses though sulfate and nitrate biases (Coutant, 1977, Meserole et al., 1979, Spicer et al., 1978) have been observed due to adsorption of sulfur and nitrogen oxide containing gases by the filter medium during sampling; the HIVOL glass fiber filter material experiences this artifact formation. Intermethod comparisons for ion chromatography (Butler et al., 1978) and automated colorimetry (Mueller et al., 1980) show them to be accurate and precise analytical methods for a number of ions.

Organic Compounds

The number of organic compounds in suspended particulate matter is high; it will probably be impractical to quantify them all. Those species important to receptor modeling will be those which are unique to certain source types and retain their chemical character between source and receptor. These compounds have not yet been identified, but a number of methods are under development for that purpose. They are all destructive and their sensitivities, accuracy and precision are unknown at the current state-of-the-art. Benzene extraction of glass-fiber filters followed by weighing of the residue has been widely used in the past to quantify the organic content of HIVOL samples, but since Grosjean (1975) showed that this solvent does not dissolve all organic materials, its use has decreased. Appel et al. (1979) propose a dual extraction on different sample sections with cyclohexane and a benzene methanol-chloroform combination; they assert that these extractions differentiate between organic material of primary and secondary origins. Daisey et al. (1979) have used ambient polynuclear aromatic hydrocarbon concentrations in receptor models. Cronn et al. (1977) have used high resolution mass spectrometry (Schuetzle et al., 1973) to identify many organic compounds in ambient aerosols. Standardized procedures, the definition of lower quantifiable limits, precision, accuracy and interlaboratory comparisons are lacking for the organic compound analysis methods.

Inorganic Compounds

X-ray diffraction analysis has been applied to suspended particulate matter samples (Davis and Cho, 1977) and can identify the types of minerals present (Davis et al., 1979). This identification is useful in distinguishing the sources of fugitive emissions. Minerals can be identified very specifically by x-ray diffraction, even to the extent of measuring crystallographic parameters and subspecies composition (such as "iron rich chlorite", etc.). Other inorganic compound analysis methods, such as ESCA, Auger Electron Spectroscopy, SIMS, Laser Raman, and IR absorption have

received limited study for aerosol analysis and could develop into useful techniques. Standardized procedures, lower quantifiable limits, precision, accuracy and interlaboratory comparisons are lacking.

Optical and Physical Properties

Microscopy, as noted earlier, can measure many properties of the aerosol by examining a number of the individual suspended particles which it comprises. Optical microscopy (McCrone and Delly, 1973, McCrone et al., 1978) is best suited to the examination of particles in the coarse mode (>2 μm optical diameter). Electron microscopy (Lee et al., 1979) can examine fine as well as coarse particles and can be automated to reduce subjectivity and analysis costs. Electron and optical microscopy standardized procedures for preparing samples, sizing particles, assigning densities, and measuring particle properties have not been developed; Crutcher (1979) has proposed procedures for optical microscopy which merit evaluation and development. Crutcher and Nishimura (1978) have also identified the sources of experimental error in microscopic analysis results as sample population statistics, mensuration methods and human factors. They propose ways to quantify these uncertainties and propagate them through the final results.

Bradway and Record (1976) and Crutcher (1978) have performed interlaboratory comparisons of optical microcopical analysis; the results cast doubt on the accuracy and precision of the method, but considering the lack of standardization among participating laboratories, these findings are not surprising. These interlaboratory tests should be undertaken again after a standard method is developed.

Development areas for analytical methods applied to receptor models should include the following:

- Critically review all aerosol analysis methods for their practicality to receptor modeling. Determine the extent to which they meet the necessary and desirable criteria. Identify potentially useful methods for development.

- Develop standard reference materials representing different ambient and source particulate matter matrices. Characterize them in terms of relevant aerosol properties.

- Standardize and conduct intermethod comparison tests of organic compound, inorganic compound, carbon, and microscopical analysis methods.

CHAPTER 4: RECEPTOR MODEL NEEDS

Receptor modeling is a new and growing field. Unlike many fields of science which start with a few key researchers and branch out to include new ones, receptor modeling's roots are spread over an array of disciplines and organizations. Nuclear spectroscopists, chemical engineers, industrial plant managers, regulatory agency personnel — each has approached the relationship of source to receptor by aerosol properties from his own point of view. It is not surprising that a coherent theory of receptor modeling and standardized methodologies have yet to emerge.

Each of the study areas summarized in the previous sections has ended with a list of research recommendations. These lists are not exhaustive, but they do contain those issues which the most experienced scientists in the field of receptor modeling believe are in need of resolution for the specific study area. For receptor modeling in general, five major development areas exist:

The primary need of receptor modeling today is a general theory within which to operate. Each specific receptor model application should be derivable from this framework. If not, either the application of the theory is incorrect, or the theory must be changed. Throgmorton and Axetell (1978) have compiled the applications. Henry (1977) and Watson (1979) have outlined the theory including some of those applications. The theory must be expanded to include them all.

Next, the applications have to be validated and placed into standardized forms. Validation should consist of two steps. First, simulated data sets of aerosol properties should be generated from pre-selected source contributions as did Watson (1979) in his simulation studies of the chemical mass balance method. These data should be perturbed with the types of uncertainties expected under field conditions. The types of sources and their contributions predicted by the receptor model application should be compared with the known source model values and the extent of perturbation tolerable should be assessed. This will define the precision required of the input data for the application in question.

The next step in validation requires a real data set with as many properties of source and receptor as can be obtained in an area with well-known sources. All receptor models should be applied and the results should be examined for a self-consistent picture of source contributions.

This consistency should include the following elements:

1. Concentrations of all measured species should be predicted to within a specified tolerance.

2. Concentrations of chemical species which originate in a source type should be highly correlated.

3. Spatial source contribution fluctuations should be extreme for point source and smooth for homogeneously distributed.

4. For samples taken during periods of constant wind direction source contributions should be higher when a source is upwind of a receptor than when it is downwind.

5. Source contributions to different particle size ranges should be consistent with the measured size distribution of the souce emissions.

If a consistent picture does not emerge, the inconsistent applications must be reformulated or discarded. A systematic elimination of measurements from the data set should then be made to find the most cost-effective sampling and analysis scheme to provide valid information within a specified confidence interval.

The third need is standardization. The receptor model applications need to be written as standard computer routines, common data structures that can accommodate uncertainties of the observables need to be created, and sampling and analysis equipment and procedures must produce equivalent results. Field study "scenarios" for typical situations should be proposed.

The fourth need is for aerosol property measurement methods. Cost-effective aerosol composition size and variability measurements at both source and receptor are required for receptor models to work. Their predictions will be only as good as the information presented to them. Though several sampling and analytical techniques have reached a high degree of sophistication, many gaps remained to be filled. The

most important of these is adequate source sampling and characterization, the lack of which appears to be the major factor in limiting advances in the state-of-the-art of receptor modeling. A systematic development of source and ambient sampling and analytical techniques must be defined, priorities must be assigned, and the work must be carried out.

The fifth and final need is for documentation and education. The validation and standardization will go for naught if the practice of receptor modeling cannot be established at the state implementation plan level where it is most sorely needed. Major reviews of model applications, analytical methods, source characterization and field study design need to be prepared and communicated to those most likely to make use of them.

The mechanisms for carrying out the research required are not clear. These mechanisms must be cost-effective while involving as wide an array of talent as possible. It is evident from the meeting of experts which generated this report that cross-disciplinary and inter-institutional approaches to meeting these needs must be fomented.

Enough receptor modeling by other names is going on today that, with the proper coordination and leadership, the cost of pursuing these developments should not be prohibitive. Most of the development should take place within the context of ongoing projects. EPA, however, needs to provide the direction and small amounts of financing to get the extra mileage out of these projects.

An EPA task force on receptor models needs to be established. It should incorporate representatives of the suspended particulate matter standards making and enforcing, ambient monitoring, and source characterization entities. This task force should keep itself informed of the status of non-attainment areas and the studies being undertaken to formulate implementation plans; its members should also keep abreast of regional air quality studies funded by EPA and industry. The task force should define the needs stated in this

report in operational terms which can be economically incorporated into the ongoing studies, arrange additional financing to cover the additional scope of work, monitor the progress toward meeting the needs, and document the results. This has been done in the past for source models. Receptor models have shown enough potential that they deserve similar treatment.

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